Process for manufacturing $(U,Pu)O_2$ mixed oxide nuclear fuel pellets from non-free-flowing UO_2 powder

The present invention relates to a process for manufacturing a $(U,Pu)O_2$ mixed powder from non-free-flowing UO_2 powders.

The manufacture of fuel for light-water reactors, based on uranium and plutonium oxides, generally called MOX fuel, has been the subject of various developments associated with the desire to to recycl plutonium recovered during spent fuel reprocessing.

The manufacture and irradiation of MOX fuel in light-water reactors are now considered to be a solution for providing reasonnable resistance to the proliferation of plutonium present in a form separated from the fission products, whether this plutonium is either of civilian or military origin.

Several processes for manufacturing MOX fuel have been developed over the last two decades, some of the processes involving the complete milling of the UO_2 and PuO_2 powders in order to provide an intimate blend, while others are limited to milling only a fraction of these powders.

The MIMAS (standing for MIcronization and MASter blend) process, which was developed by the Applicant of the present invention (see figure 1), comprises the micronization, by milling, of only a fraction of the final blend and uses two successive blending operations to achieve isotopic homogenization and to take advantage of the use of free-flowing UO_2 incoming products (especially to ensure that the dies of the presses used for pelletizing are properly filled). Using free-flowing UO_2 powders in the second blending operation and limiting the milling to only the first blending operation simplify the manufacture (for example by dispensing with the operations of precompacting/granulating or spheroidization of the

mixed oxide blend) and have greatly simplified, at the start of industrial implementation , the qualification of MOX fuel by users and the licensing process by the Nuclear Safety Authorities (thanks to the similarity in behavior between this MOX fuel and UO_2 fuel).

Various versions of the MIMAS process have been applied, sometimes under names different from MIMAS, but all characterized by two successive blending operations, the second of which uses free-flowing UO_2.

UO_2 which serves as feed material in the manufacture of enriched-uranium fuel and, in the great majority of cases, in the manufacture of MOX fuel, is obtained by the conversion of uranium hexafluoride. There are industrial conversion processes which produce free-flowing UO_2 powder. This is especially the case with two industrial conversion processes using a wet route, known in the art by the respective names "AUC", coming from the intermediate product (Ammonium Uranyl Carbonate), and "TU2", coming from the uranium transformation unit in which the conversion is carried out. One of the drawbacks of these wet conversion processes is the production of a large amount of liquid effluents which have to be treated before discharge. The wet conversion processes, some of which do not produce free-flowing UO_2, are gradually being replaced with dry processes which allow the gaseous effluents to be recycled but which generally produce non-free-flowing UO_2 powder.

For the purpose of diversifying the sources of UO_2 powder for manufacturing MOX fuel by MIMAS-type processes, it is therefore useful to be able to employ non-free-flowing UO_2 powders.

Non-free-flowing UO_2 powder conditioning processes, for transforming it into free-flowing UO_2 granules, and therefore having properties suitable for feeding a pelletizing press, are known. Various mechanical granulation processes, such as precompaction-granulation or agglomeration-

spheroidization, have been developed and are used on an industrial scale in UO_2 fuel manufacturing plants.

Experience has shown that these granulation processes produce granules of insufficient mechanical strength for correct implementation of the second blending operation which characterizes the MIMAS processes and similar processes. Under the optimum operation of the second blender, the granules are damaged and the flowability of the secondary blend is impaired: the fuel pellets which result therefrom suffer from major defects (excessive variability in the physical properties of the product, local differential shrinkage defects, etc.). Alternatively, if the method of operating the second blender is modified so as to achieve gentle mixing of the powders to be blended, or if the apparatus used for the second blending is modified for the same purpose, the uniformity of distribution of the plutonium within the fuel may be impaired and the MOX pellets thus produced no longer meet the maximum plutonium content variability criteria.

To avoid the abovementioned drawbacks, the process for manufacturing MOX fuel from non-free-flowing UO_2 powder, which is the subject matter of the invention, comprises a mechanical granulation treatment of the non-free-flowing UO_2 powder, which does not modify the chemical properties (such as a stoichiometry) and morphological properties (such as the particle size) of the UO_2 powder, but which does nevertheless ensure the mechanical strength and flowability that are required to successfully carry out the second blending operation and the pelletizing operation, respectively.

The invention thus obviates the need to supply the MIMAS-type processes with free-flowing UO_2 powders as feed materials.

According to one advantageous method of implementing the invention, non-free-flowing UO_2 powder is used, one part of which is used, as it is, for

incorporation in the first blend and one part of which undergoes a granulation treatment before being incorporated into the second blend.

In a variant, as a nonlimiting example, said granulation treatment may also be applied to the non-free-flowing UO_2 fraction fed in the first blend

In order to avoid the drawback of the abovementioned lack of mechanical strength of UO_2 granulated by one of the usual conditioning processes, the mechanical treatment according to the invention is carried out either by forcing the non-free-flowing UO_2 powder through a screen or sieve, or by compressing this powder into tablets under a high pressure, as required for obtaining suitable non-friability properties, and then crushing said tablets. When necessary, one or more binders and/or lubricants may be added beforehand to the UO_2 powder.

Further details and features of the invention will become apparent from the claims and from the description of the drawings, which are appended to the present specification and which illustrate, by way of nonlimiting examples, the manufacturing process according to the invention.

Figure 1 shows schematically the steps in the manufacture of mixed oxide fuel according to a known process of the MIMAS type.

Figure 2 shows schematically the steps in the manufacture of mixed oxide fuel according to a process of the invention.

Figure 3 shows schematically variants of the process according to the invention.

In the various figures, the same reference notations denote identical or similar components.

The process of the invention, for the use of non-free-flowing UO_2 powder, comprises basically a process for the manufacture of $(U,Pu)O_2$ mixed oxide fuel pellets, that is to say overall (figure 2):

- dosing and first blending (step 1) of PuO_2 powders and/or UO_2 powders and/or fuel manufacturing scrap;
- micronization (step 2) of this first blend, particularly by milling, and forced sieving (step 3) of its product, for example through a 250 μm screen mesh;
- additional dosing and second blending (step 4) of the first blend thus treated, UO_2 and, where appropriate, fuel manufacturing scrap;
- addition, and blending with the resulting second blend of one or more lubricants and/or poreformers (step 5), the latter step possibly being completely or partly combined with step 4;
- compression (step 6) of the second blend into pellets using pelletizing presses ; and
- sintering (step 7) of the pellets thus formed, preferably in an atmosphere of moistened argon (or nitrogen) and hydrogen.

This mixed oxide fuel pellet manufacturing process may also usually include, for the pellets thus obtained, steps of:

- dry grinding (step 8);
- visual inspection (step 9);
- stacking up to length (step 10);
- loading the pellets into a cladding and welding the latter so as to form a fuel rod (step 11, figure 1);
- pressurizing the rods;
- nondestructive testing/examination of the rods (step 12); and
- assembling of the rods (step 13).

Said process of the invention furthermore includes (figure 2) a prior mechanical granulation treatment of all or part of the nonflowing UO_2 (step 29). This treatment may comprise, for example:

- either (figure 3) steps of compressing the non-free-flowing UO_2 into tablets (step 30) and of

crushing these tablets (step 31) and, where appropriate, of sieving the crushed material (step 32) in order to form free-flowing granules having properties suitable for being incorporated as the basic constituent in the second blending operation (step 4) or, in a variant, in both blending operations (steps 1 and 4), while maintaining the original chemical composition and original particle size of the original UO_2;

- or an agglomeration/precompaction/granulation step by forcing the non-free-flowing UO_2 powder through a screen or sieve (step 29), the amount of additive(s), the mesh size of the screen or sieve and the pressure exerted on the powder being adjusted in order to form granules having the suitable properties described above.

A few nonlimiting parameters of the pellet manufacturing process are given below by way of example:

- batch/campaign operation rather than continuous operation;
- plutonium content of the first blend: 20 to 40% (step 1);
- milling (step 4) in 60 kg batches for a minimum effective time of 5 hours;
- use of non-free-flowing UO_2 powders coming from a wet conversion (for example, ex-ADU or ammonium diuranate powder) or from a dry conversion (said conversions being known to those skilled in the art);
- addition of 0.2 to 0.5% of zinc stearate and 0 to 1% of an AZB pore former (known to those skilled in the art);
- pelletizing compression (step 6) at a pressure between 400 and 700 MPa;
- sintering (step 7) for at least 4 hours at a temperature between 1600 and 1760°C, in an

argon atmosphere containing 5% hydrogen, with an H_2/H_2O ratio of 10 to 30; and

- dry centerless grinding (step 8).

By way of nonlimiting example, the compression step (step 30) may be carried out at a pressure of between 50 and 200 MPa, this being tailored according to the characteristics of the non-free-flowing powder. These pressures are therefore higher than the granulation pressures (4 to 10 MPa) generally used in UO_2 nuclear fuel manufacturing plants. Some binder and/or lubricant, both well known to those skilled in the art, may be incorporated into the non-free-flowing UO_2 powder before compression: by way of nonlimiting example, the compression may thus be carried out at a pressure of between 40 and 100 MPa.

Also by way of nonlimiting example, the aforementioned tablets may be crushed in one or more jaw crushes or roll mills of 200-250 μm aperture. This crushing may be followed by sieving if the crusher lets through, or runs the risk of letting through, granules having a size greater than 250 μm. The fines possibly resulting from the crushing may usefully be incorporated as raw material into the first blending operation (step 1).

By way of yet another nonlimiting example, the operation of forcing the powder through a sieve (step 29) may be carried out in a machine of the kind used in MIMAS-type processes (step 3) to fill the first blend (after the micronization of step 2) before the second blending (step 4). Such machines, which combine agglomeration/precompaction upstream of the sieve and control of the maximum granule size by passing the powder through this same sieve, may produce granules of the desired characteristics directly.

Experience has shown the Applicant that a non-free-flowing powder treated according to the process forming the subject matter of the invention can be used in existing MOX manufacturing plants, by adjusting the parameters of this second blending operation (step 4),

the pelletizing (step 6) and the sintering (step 7), within the adjustment limits routinely used to optimize the manufacturing process according to the characteristics of the various free-flowing UO_2 powders used for MOX fuel manufacture.

The process of the invention therefore makes it possible to extend the range of UO_2 powders which can be used to manufacture MOX fuel, without loosing the benefit of the similarity between the MOX fuel produced according to the invention and the UO_2 fuel manufactured on an industrial scale by the processes known hitherto, starting from the same non-free-flowing UO_2 powder.

It should be understood that the present invention is in no way limited to the methods of implementation described above and that many modifications may be made thereto without departing from the scope of the claims given hereafter.

The non-free-flowing UO_2 conditioning process may especially be applied to UO_2 coming from a conversion other than the conversion of uranium hexafluoride into UO_2.